UCRL-93186
PREPRINT

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This paper was prepared for submittal to
Advances in Cryogenic Engineering
CEC/ICMC
Massachusetts Institute of Technology
Cambridge, MA August 12-16, 1985

August 1985

Lawrence Livermore National Laboratory

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ABSTRACT

Three different neutron sources were used to irradiate identical sets of NbTi superconductors up to about half the lifetime dose of a superconducting magnet in a fusion reactor. Based on a careful source characterization of the TRIGA Mark-II reactor in Vienna, the spallation neutron source IPNS at Argonne and the 14 MeV neutron source RTNS-II at Livermore, the damage energy cross sections were calculated for four different types of NbTi alloys (42, 46.5, 49 and 54 wt% Ti). The experimental results on the variations of critical current densities j_c with neutron dose are found to scale within the experimental uncertainties with the appropriate damage energy cross sections. This first explicit proof of damage energy scaling for j_c-variations in superconductors is considered to be most valuable for the evaluation of radiation damage in superconductors under fusion reactor conditions.

INTRODUCTION

The primary objective of this work was to determine the effects of neutron irradiation obtained from different sources on the critical current density j_c in commercially manufactured NbTi superconductors suited for magnet construction in future fusion reactors. The crucial question raised by magnet designers, whether the results from various irradiation experiments obtained so far can be directly applied to the superconductor at the magnet location, suggested the experimental verification of the damage energy concept developed in recent years. This method is increasingly used to characterize and compare the results of different neutron irradiation studies.

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The changes of the critical parameters of a superconductor such as T_c, H_{c2} and j_c during irradiation is primarily due to the total damage deposited in the material. The calculation of this quantity, measured in displacements per atom (dpa) or more practically eV/atom, requires a complete knowledge of the neutron spectrum, the fluence obtained during irradiation, the differential neutron cross sections and the primary recoil distribution of the irradiated material. In metals, a significant contribution to the total damage is caused by high energy neutrons; hence, the flux of several irradiation facilities is frequently quoted for energies greater than 0.1 or greater than 1.0 MeV, respectively. A large fraction of the incident neutron energy goes into electronic excitation, activation and thermal vibrations of the lattice atoms rather than into production of displacement cascades. Moreover, the majority of produced defects, may they be point defects or entire cascades, are subject to recombination during the process of formation, thus the temperature maintained during irradiation affects the number of "surviving" defects. An adequate model of the interaction between neutrons and the atoms of the irradiated metal has been discussed previously.

Based on the knowledge of differential elastic and inelastic neutron scattering cross sections,[3] $\sigma(E)$, the primary recoil energy distribution $T(E)$ for each individual energy-flux group of a given neutron spectrum has been calculated using the SPECTER-computer code.[4] The total displacement energy cross section $\langle\sigma \cdot T\rangle$ averaged over all neutron energies can be expressed as

$$\frac{\int \sigma(E) \cdot T(E) \cdot \frac{d\phi}{dE}\, dE}{\int \frac{d\phi}{dE}\, dE} \tag{1}$$

Multiplication of this quantity with the actual fluence obtained during irradiation yields the average energy per atom deposited for defect production. As indicated earlier, the fluence (flux ϕ x irrradiation time t) may be specified for $E > 0.1$ or $E > 1.0$ MeV, respectively. The resulting product, however, remains constant for the damage energy cross sections are scaled accordingly.

If there is more than one element in the irradiated sample, which is the case in the investigated NbTi alloys, one has to calculate the contribution of each element to the total damage energy cross section. Linear scaling with the atomic percentage c_i is employed during summation:

$$\underset{\text{Alloy}}{\langle\sigma \cdot T\rangle} = \sum_{\text{Elements}} \langle\sigma \cdot T\rangle_i \cdot c_i \tag{2}$$

More advanced theoretical models for binary systems[5] indicate that in the present case, this simplification does not represent a significant source of error. All irradiations of the NbTi superconductors described in this work have been carried out at room temperature. Irradiation experiments at 5.0 K with a subsequent annealing cycle to 300 K have been completed and will be published.[6]

EXPERIMENT

Materials

All NbTi conductors were manufactured by Brown Boveri & CIE/Switzerland. Two identical sets containing 14 single core conductors and three multifilamentary conductors have been selected for the irradiation programs at

IPNS/Argonne and RTNS-II/Livermore. A detailed characterization of all irradiated materials is given in Ref. 7. One of the multifilamentary conductors has been developed for the LCT (Large Coil Test) program currently carried out at Oak Ridge National Laboratory. An additional set has been irradiated earlier in the TRIGA Mark-II reactor in Vienna.[7] All single core conductors have undergone an annealing step at 400°C prior to final cold work which was varied (0 - 91%) as well as the Ti concentration (42, 49 and 54 wt% corresponding to 58, 65 and 70 at% respectively). Whereas the Ti-rich samples (54 wt% Ti) exhibit significantly higher current densities in manetic fields up to 6 T than materials with 42 wt% Ti, their performance at the projected field of 8 T is declining rapidly because of their lower H_{c2}. For the present standard conductors, including the LCT-conductor, a composition of 46.5 wt% (63 at%) Ti has been chosen. The single core conductors were all 0.4 mm in diameter, their Copper to superconductor ratio (with respect to volume) was about 1:1. All samples were prepared to meet the requirements of the experimental setup measuring j_c and the spacial constraints in the irradiation facilities. Straight pieces, 47 mm in length, were cut and polished to remove the oxide layer from the copper stabilizer. Thin potential leads were soldered onto each sample with a relatively small gauge length of 6 mm to avoid excessive fluence gradients along the sample arising from the strong inhomogenious flux distribution inherent to all accelerator based neutron sources.

Neutron Irradiation

One set of samples has been irradiated with 14.6 MeV neutrons at RTNS-II in 3 cycles to fluences of 2.22×10^{21}, 4.26×10^{21} and 5.82×10^{21} n/m^2 averaged over all specimens. Small Nb foils attached to the samples were used to monitor the fluences. After each cycle the whole batch was shipped to Argonne for j_c-measurements. Before the second and third cycle the positions of the samples have been rotated with respect to the beam spot of the target in order to compensate the differences in neutron fluence between individual samples.

Another set has been irradiated at ambient temperature at IPNS in 3 cycles up to fluences ($E > 0.1$ MeV) of 0.53×10^{22}, 0.86×10^{22} and 1.96×10^{22} n/m^2 with spallation neutrons. These numbers are based on an activation analysis of Ni dosimetry wires attached to the samples during irradiation.

The results on an identical set of materials irradiated in the TRIGA-reactor were already reported by Nardai et al.[7] and are used for comparison. Although there have been six irradiation cycles carried out up to a total fluence of about 1.75×10^{23} n/m^2, only data for the first three cycles, 8.8×10^{20}, 8.8×10^{21} and 4.4×10^{22} n/m^2 ($E > 0.1$ MeV), are quoted in this work because the results at higher fluences exceed the range of data obtained from the experiments at RTNS-II and IPNS. It should be noted that the absolute errors in determining the neutron fluence are typically estimated at 10%.

j_c-Measurements

After each irradiation cycle the critical current densities have been measured at 4.2 K as a functiton of the magnetic field (perpendicular to the wire axis) in the range between 1.0 and 8.0 T in steps of 0.5 T. Each sample was mounted free from mechanical stress onto a sample holder in a manner to prevent any movement caused by the Lorentz force when magnetic fields were present during j_c measurement. A standard four probe technique was employed to determine j_c applying a voltage criterion of 50 μV/cm. The implications of "defining" j_c by means of a certain voltage criterion have been discussed earlier.[8] In the case of single core conductors, this does not represent a major problem for their transition from the superconducting to the normal state occurs quite rapidly in contrast to multifilamentary conductors with a rather smooth I-V characteristic near j_c.

ANALYSIS

Neutron spectra

Based on elastic and inelastic scattering cross sections, i.e. nuclear data for (n,xn), (n,α) and (n, γ), reactions listed in ENDF-B/V,[3] the differential fluxes $d\phi/dE$ in all 100 energy groups have been calculated using the STAYS'L computer code.[9] An activation analysis[10] of the central irradiation thimble of the TRIGA Mark-II reactor in Vienna with subsequent STAYS'L evaluation revealed no significant deviation from the spectrum originally specified by the contractor General Atomics/San Diego. The neutron spectra of the irradiation facilities used are illustrated in Fig. 1. where the flux per unit lethargy, $d\phi/d\ln E$, is plotted on a linear scale. In this representation the area under the spectrum or sections thereof is directly proportional to the total number of neutrons or its fraction within a specified energy range.

In characterizing the neutron spectra of the three facilities, one has to deal with different source strengths which are summarized in Table 1. The spectra have been normalized in a way that the total of all group fluxes is equal to 1.0 which is useful if one is comparing the percentage of neutrons within a particular energy range between different neutron sources regardless of their total flux. In addition, the theoretically modelled spectra at the magnet location for the STARFIRE[11] and MARS (Mirror Advanced Reactor Study)[12] designs are presented in Fig. 2., viewing the regions with the least amount of shielding between the plasma and the superconductor, which can be regarded as a worst case assumption.

Damage energy cross sections averaged over the entire neutron spectrum or scaled to E > 0.1 or E > 1.0 MeV respectively, have been calculated for pure Nb, Ti and several alloys. The numbers listed in Table 2. are applicable to fluences quoted for E > 0.1 MeV. The damage energy cross sections

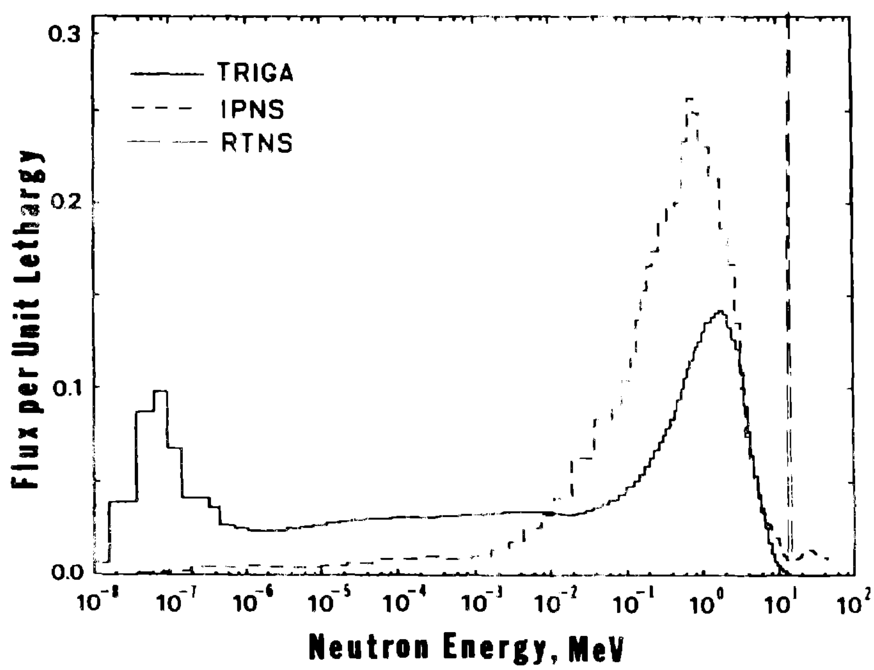

Fig. 1. Spectra of the neutron sources used during the irradiations.

Table 1. Flux densities of different neutron sources and calculated fusion reactor designs, Units in $n/m^2 \cdot s$.

Source/ design	Total	E > 0.1 MeV	E > 1.0 MeV
TRIGA-reactor, Vienna[a]	$1.9x10^{17}$	$7.5x10^{16}$ (38.8%)	$3.9x10^{16}$ (20.3%)
IPNS/REF[b]	$1.0x10^{16}$	$6.9x10^{15}$ (71.3%)	$2.7x10^{15}$ (27.5%)
RTNS-II[c]	----------$1.15x10^{16}$ of 14.6 MeV neutrons----------		
MARS[d]	$1.2x10^{14}$	$6.0x10^{13}$ (50.0%)	$1.1x10^{13}$ (9.0%)
STARFIRE[e]	$3.1x10^{13}$	$1.7x10^{13}$ (55.0%)	$5.0x10^{12}$ (16.0%)

[a]At 250 kW reactor power.
[b]At a beam current of 8 μA of 450 MeV protons.
[c]At beam spot under operation with 150 mA d^+-beam, 380 keV.
[d]In Yin-Yang region assuming 4.3 MW/m^2 neutron wall loading at blanket of center cell.
[e]Assuming 3.6 MW/m^2 neutron wall loading.

for both fusion reactor designs are found to be slightly smaller than those of the TRIGA-spectrum and are very close to the numbers obtained for IPNS.

Results

The results of all j_c-measurements prior and after the irradiations have been recorded and are available in files under VAX/VMS. The selection of data presented here primarily emphasizes the validity of damage energy scaling and also indicates the effect of neutron irradiation on materials with different Ti-concentration and degree of final cold work (fcw). A more detailed analysis is given in Ref. 6.

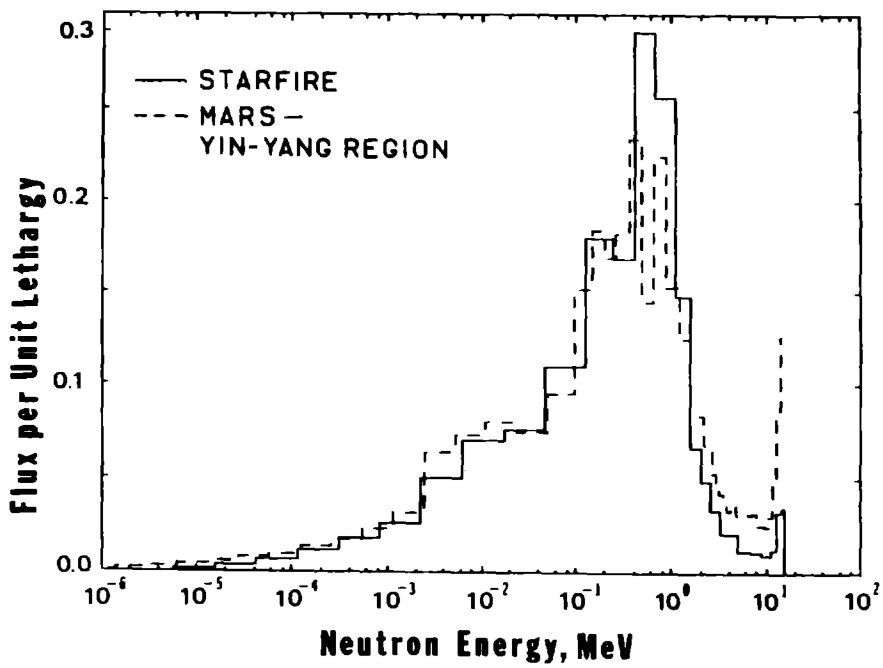

Fig. 2. Calculated neutron spectra at the magnet location for two fusion reactor designs.

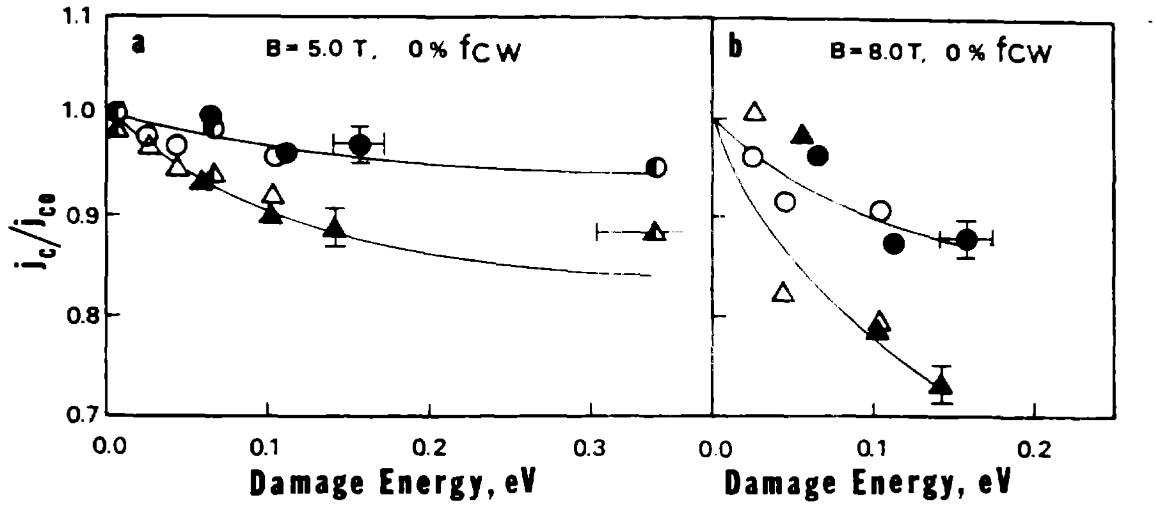

Fig. 3a. Fractional change of the critical current density j_c at 4.2 K and 5.0 T as a function of the deposited damage energy for Nb-42 wt% Ti(○◐●) and Nb-54 wt% Ti (△◭▲) with no fcw. The light symbols represent the results of the irradiation at IPNS/REF, the shaded ones those of the TRIGA-reactor, and the bold ones those of the 14 MeV neutron irradiation at RTNS-II

3b. Equivalent to Fig. 3a. but for B = 8.0 T.

The fractional changes of j_c as a function of the damage energy at 4.2 K and 5.0 T for all 3 irradiations are illustrated in Fig. 3a. The results for both Nb-42 wt% Ti (○◐●) and Nb-54 wt% Ti (△◭▲) fit a smooth line within the experimental uncertainties of the dosimetry results (+-10%) and of the quantity j_c/j_{co} which depends slightly on the absolute value of the critical current I_c (typically 2%). The reasons for the observed differences in j_c-degradation between Nb-42 wt% Ti and Nb-54 wt% Ti are not completely understood at this time, particularly in consideration of the lacking final cold work in these materials which is required for a well defined microstructure and efficient flux pinning. As displayeed in Fig. 3b., this trend was found to be enhanced by a factor of about 3.0 at higher fields (8.0 T) where the performance of Nb-54 wt% Ti dropped to 70% of its value prior to irradiation. We assume that a gradual change of the upper critical field H_{c2} during neutron irradiation is responsible. H_{c2} has been measured prior to irradiation, corresponding post-irradiation data are still awaited

Table 2. Displacement damage energy cross sections $\langle\sigma \cdot T\rangle_E > 0.1$ MeV for different neutron sources and calculated fusion reactor designs, units in keV barn

Source Design	Ti	Nb	Nb-42wt%Ti	Nb-46.5wt%Ti	Nb-49wt%Ti	Nb-54wt%Ti
TRIGA-reactor,						
Vienna	81.5	70.7	76.9	77.4	77.7	78.2
IPNS/REF	64.9	62.0	63.7	63.8	63.9	64.1
RTNS-II	248.5	280.4	261.9	260.3	259.7	258.1
MARS,						
Yin-Yang region	58.3	62.9	60.3	60.1	59.9	59.7
STARFIRE	66.5	67.9	67.0	67.0	67.0	66.8

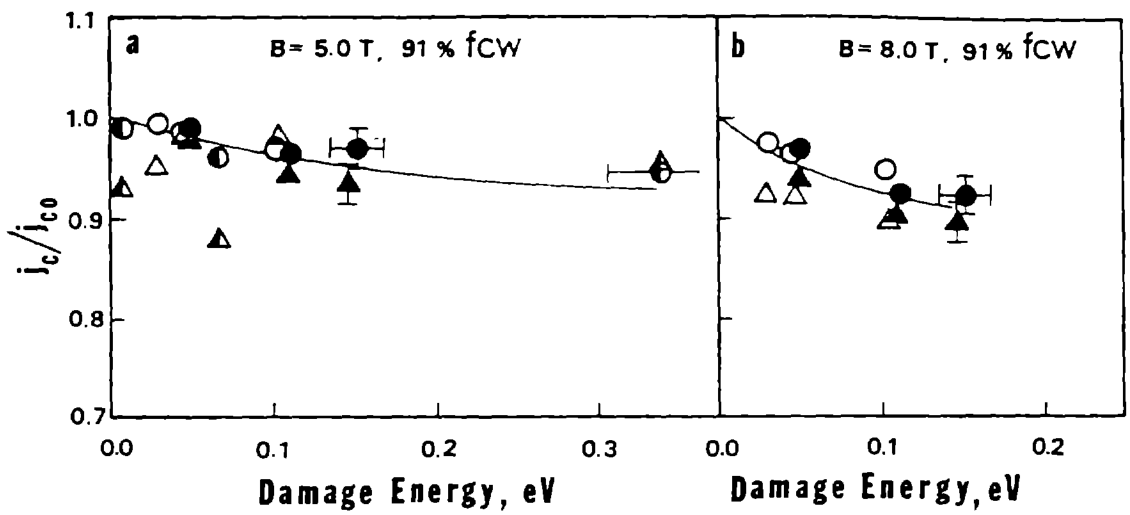

Fig. 4a. Fractional change of the critical current density j_c at 4.2 K and 5.0 T as a functin of the deposited damage energy for heavily cold worked Nb-42 wt% Ti(○◐●) and Nb-54 wt% Ti (△▲▲) the light symbols exhibit the results of the irradiation at IPNS/REF, the shaded ones those of the TRIGA-reactor and the bold ones those of the 14 MeV neutron irradiation at RTNS-II.

4b. Equivalent to Fig. 4a. but for B = 8.0 T.

In contrast, the response to neutron irradiation of heavily cold worked superconductors depicted in Figs. 4a. and 4b. is significantly smaller. Apparently their structure with a high density of dislocation cells and cell walls which is primarily responsible for an improvement in the flux pinnning mechanism and consequently in j_c, is equally affected by irradiation in both materials. Considering that the absolute increase in the normal state resistivity ρ_o is roughly equal in both cold worked (Figs. 4a. and 4b.) and non-cold worked NbTi (Figs. 3a. and 3b.), one expects higher j_c-degradiation in materials with 0% fcw. The change of j_c can be written as

$$\frac{\Delta j_c}{j_{co}} = -c_1 \frac{\Delta\rho}{\rho_o} + c_2 \frac{\Delta H_{c2}}{H_{c2}} + c_3 \frac{\Delta T_c}{T_c} \tag{3}$$

with j_{co}, ρ_o, H_{c2} and T_c representing the values prior to irradiation. Our experimental observations demonstrate that NbTi with 0% fcw and initially lower ρ_o are more affected by irradiation. A more detailed analysis of the effects of neutron irradiation with respect to the metallurgical parameters of NbTi superconductors will be presented later. For practical applications, the absolute value of j_c prior to irradiation is as important as their performance during neutron irradiation. In cold worked (91%) Nb-54 wt% Ti, j_c was found to be 1.7×10^9 A/m^2 at 5.0 T and 0.61×10^9 A/m^2 at 8.0 T, whereas the same material with 0% fcw yielded 8.3×10^8 and 2.2×10^8 A/m^2 respectively.

As further evidence for the validity of damage energy scaling, we present the results on the Swiss LCT-conductor in Fig. 5. At 8.0 T, the highest magnetic field in which NbTi superconductors are expected to be employed in projected magnet designs for fusion reactors, its performance decreases roughly 10% after half the lifetime fluence.

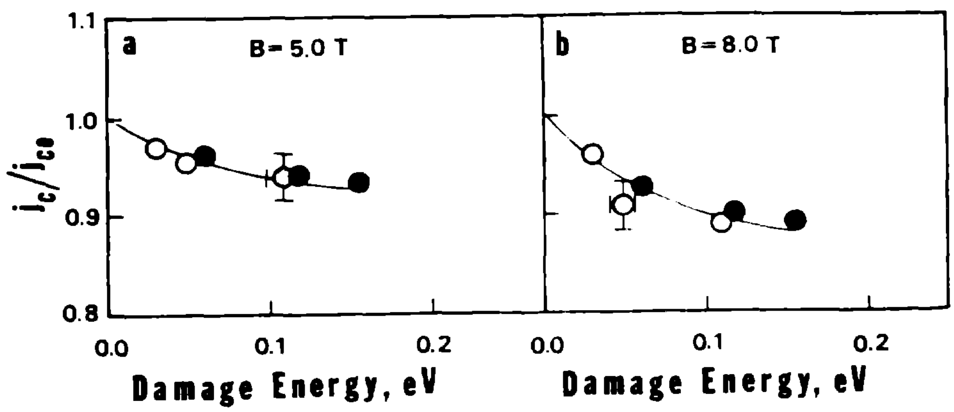

Fig. 5. Fractional change of j_c at 4.2 K as a function of the deposited damage energy for the Swiss LCT-conductor. The light symbols represent the result of the irradiation at IPNS/REF, the bold ones those of the 14 MeV irradiation at RTNS.

CONCLUSIONS

In general, the results of all three irradiation experiments on NbTi superconductors were found to agree when the obtained fluences are scaled with the appropriate damage energy cross sections. This experimental observation enables magnet designers to precisely determine the effects of neutron irradiation in magnet components since the effects of a fusion spectrum can be inferred from irradiations in other spectra. Considering technological applications of NbTi on a large scale, materials with multiple thermomechanical treatment are suited best for they have both higher initial critical current density and are least effected by irradiation.

ACKNOWLEDGEMENTS

The authors are gratefully indebted to the technical staff of IPNS and RTNS-II for their support during the experiments and Ruth M. Nuckolls for handling the dosimetry. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48 and by the Federal Ministry of Science and Research, Wien, Austria.

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